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④ Method of producing level off DP microcrystalline cellulose and glucose from lignocellulosic material.

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**ABSTRACTS BULLETIN OF THE INSTITUTE OF
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Courier Press, Leamington Spa, England.

Description

This invention relates to a method of producing level off DP microcrystalline cellulose and glucose from lignocellulosic material.

More particularly, this invention relates to the use of an explosive depressurization reactor for the production of level off DP microcrystalline cellulose and glucose from lignocellulosic material. Until my inventions to render lignin separable from cellulose and hemicellulose and the product so produced (Canadian Patents 1,096,374 and 1,141,376), there was no known economically viable process to cleanly separate undegraded and chemically reactive lignin and the hemicellulose from the cellulose in lignocellulosic material. Thus, until now, acid hydrolysis of lignocellulosic biomass has been done by treating the material as a composite.

In this specification, "lignocellulosic material" includes such plant growth materials as oat hulls, corn stalks, bagasse, wheat straw, oat straw, barley straw and woods of various species, particularly hardwoods. Lignocellulosic material is comprised of three main chemical components—lignin, hemicellulose and cellulose—in the following approximate proportions, plus ash and trace elements:

Hardwoods:	
Lignin	21%
Hemicellulose	24%
Cellulose	48%

Annual Plant Material (Straw, Bagasse, etc.)	
Lignin	15%
Hemicellulose	31%
Cellulose	46%

The cellulose and hemicellulose are both carbohydrates. Cellulose is nature's most abundant chemical, hemicellulose is second and lignin is third. Cellulose is comprised of six-carbon (glucose) sugar molecules. The xylan component (approximately 70%) of the hemicellulose in annuals and hardwoods is comprised mainly of five-carbon (xylose) sugar molecules. The lignin is a complex amorphous molecule comprised of many of the chemical components found in oil and gas such as phenol, benzene, propane, etc. The function of these materials in the lignocellulosic complex is as follows:—

The core of the lignocellulosic fibre is comprised primarily of cellulose. Cellulose is the skeleton in the figure structure. It occurs as crystalline bundles which support the fabric of the tree or plant.

The hemicellulose and lignin are cross-linked to form a matrix which surrounds the cellulose skeleton and hold the structure together in the manner of resin in a fibreglass composite.

It is this lignin/hemicellulose matrix which provides nature's protection against microbial invasion. It also renders the material water resistant.

Abstracts Bulletin of the Institute of Paper

Chemistry Vol. 47, No. 10, April 1977 p. 1095 No. 10612 (u) and FR—A—2472016 disclose methods of hydrolysing cellulose utilising sulphuric acid. EP—A—44658 and FR—A—1007264 disclose methods of hydrolysing cellulose utilising high-pressure steam.

With the application of the teachings of Canadian Patents 1096374 (equivalent to GB—A—2000822) and 1141376 the problem, which has baffled scientists and engineers for more than a century, of how to break the intermolecular cross-links between the lignin and the hemicellulose without causing substantial degradation of either of these chemical components has been solved. Once the intermolecular cross-links in the lignocellulosic material have been severed, it is relatively simple to partition the material into its three main chemical components (lignin, hemicellulose and cellulose) using mild organic solvents or weak sodium hydroxide.

Thus GB—A—2000822 discloses a method of dissociating lignocellulosic material, comprising: a) packing the lignocellulosic material in a divided, exposed, moist form in a pressure vessel having a valved outlet; b) with the valve closed, rapidly filling the pressure vessel with high pressure to bring, by means of the pressurised steam, substantially all of the lignocellulosic material to a predetermined temperature in the range 185 to 240°C in less than 60 seconds; c) as soon as the said predetermined temperature has been attained, opening the valved outlet and explosively expelling the lignocellulosic material in a plastic condition from the pressure vessel through the outlet to atmosphere so that the said material issues from the outlet in particulate form and; d) separating the cellulose in the particulate material from the lignin and hemicellulose.

In one variant of this method the lignin is extracted from the mixture using a mild organic solvent such as ethanol, methanol or a weak sodium hydroxide solution at room temperature, and then after filtering the remaining material is separated into cellulose and hemicellulose fractions by dissolving the hemicellulose in a weak (1 wt.%) solution of sodium hydroxide at 50—100°C, the selected temperature depending on the desired extraction time.

Alternatively the hemicellulose is extracted from the whole exploded material with warm (50°C) water for two hours, then after filtering, the lignin is extracted using a mild organic solvent such as ethanol, methanol or a mild (0.1N) solution of sodium hydroxide at room temperature, which after filtering leaves a high purity cellulose fraction.

The resulting fractions (chemical components) are very pure and largely in so-called native or undegraded form. The lignin is amorphous and very delicate chemically when it is isolated from the hemicellulose matrix. It will hydrolyse easily in a mild acid solution at or below its glass transition temperature around 125°C, the required temperature depending on moisture content:

Similarly, the xylan component of the hemicellulose is also amorphous and only slightly more rugged chemically than the lignin. It will hydrolyse quite easily in a mild acid solution at its glass transition temperature at or around 165°C, also dependent on moisture content. The crystalline component of the cellulose, on the other hand, is more rugged chemically and significantly more difficult to hydrolyse than the other two, but it will hydrolyse quite readily in a mild acid solution at or slightly above its glass transition temperature of 234°C.

When lignocellulosic material which has not been subject to the explosion process is acid hydrolysed as a composite, the acid reacts initially with the more delicate amorphous (lignin and xylan) components. Before the crystalline cellulose is hydrolysed, they are degraded beyond economic value as a chemical feedstock to what is commonly called a black liquor, and they mix intimately with the glucose, creating toxicity and a difficult separation problem. Moreover, because the microstructure of the composite is resistant to water and other liquids, the wood chip or other untreated feedstock will resist the penetration of the acid and will therefore tend to hydrolyse from the outside of the chip toward the centre. Thus, the glucose which is first produced from the outside of the chip is degraded before the glucose from the inside of the chip is released. This is the fundamental reason why yields of glucose by acid hydrolysis of lignocellulosic materials have until now been limited to below 50% of the theoretical yield.

A cellulose microfibril is made up of a cellulose microcrystal surrounded by a hinged to the next microcrystal by an amorphous region of cellulose. It has now been found that this amorphous cellulose will hydrolyse to glucose under less harsh hydrolysis conditions of time, temperature, and acid concentration than is required to hydrolyse the microcrystal to glucose. For this reason, under the proper hydrolysis conditions, it has now been found that it is possible to produce microcrystalline cellulose in a glucose solution.

It has now been found that when the methods described in GB—A—2000822 (=CA 1096374) and CA 1141376 are applied to lignocellulosic materials and the cellulose is separated from the resulting product, the cellulose is in the form of cellulose fibrils which are small in size (20—50 micrometers in diameter, and one or two millimeters in length). These fibrils have been expanded by the mechanical action of the explosive decompression, leaving voids where the lignin and hemicellulose once resided and where acid selected from the group consisting of sulfuric acid, hydrochloric acid and sulfurous acid can easily penetrate. It has further been found that when the cellulose is damp with moisture, an acid pre-soak will wick and thereby distribute the acid evenly onto each molecule of cellulose throughout the material. By this means the problem of accessibility of the cellulose to the acid and the problem of achieving an even distribution of acid throughout the cellulose are solved.

Accordingly the invention provides a method of dissociating lignocellulosic material, comprising:
 a) packing the lignocellulosic material in a divided, exposed, moist form in a pressure vessel having a valved outlet; b) with the valve closed, rapidly filling the pressure vessel with steam at high pressure to bring, by means of the pressurised steam, substantially all of the lignocellulosic material to a predetermined temperature in the range 185 to 240°C in less than 60 seconds; c) as soon as the said predetermined temperature has been attained, opening the valved outlet and explosively expelling the lignocellulosic material in a plastic condition from the pressure vessel through the outlet to atmosphere so that the said material issues from the outlet in particulate form and; d) separating the cellulose in the particulate material from the lignin and hemicellulose, characterised by said high pressure steam being at a pressure of 2.76 to 4.82 MPa (400 to 700 psi) and by the further steps of: e) soaking the separated cellulose in an acid solution of at least one acid selected from the group consisting of sulfuric acid, hydrochloric acid and sulfurous acid until the acid is distributed evenly throughout the cellulose, and then reducing the moisture content of the cellulose, leaving the cellulose impregnated with acid at a concentration in the range 0.05% to 2.0% of the weight of the cellulose; f) packing the acid-impregnated cellulose into a pressure vessel having a valved outlet; g) rapidly filling the pressure vessel with steam at a pressure in the range of 2.07 to 4.82 MPa (300 psi to 700 psi) to bring, by means of the pressurised steam, substantially all of the acid-impregnated cellulose to a predetermined temperature in the range of 185—240°C in less than 60 seconds; h) as soon as the said predetermined temperature is attained, opening the valved outlet, and explosively expelling the acid-impregnated cellulose from the pressure vessel through the outlet to atmospheric pressure; i) neutralising the resulting solution.

In some embodiments of the present invention the moisture content of the acid-impregnated cellulose may be reduced to a level in the range 20% to 80% by weight of the cellulose. This may be achieved by mechanical dewatering. Dewatering facilitates the heating of the cellulose by the steam and prevents the presence of excessive amounts of water in the pressure vessel. Preferably, the moisture content is reduced as much as is reasonably possible. The dewatered acid-impregnated cellulose may then be loaded into the pressure vessel, charged with high pressure steam at pressures between 2.07 to 4.82 MPa (300 psi and 700 psi), depending on moisture content, to bring the material to a temperature between 185 and 240°C in less than 60 seconds, preferably less than 45 seconds depending on the temperature and acid concentration. When the desired temperature is reached the acid-impregnated cellulose is explosively expelled to the atmosphere. Preferably, any residual cellulose is filtered from the glucose solution.

In some embodiments of the present invention

wherein the neutralised end product is a mixture of glucose from the amorphous component of the cellulose and level off DP microcrystalline cellulose from the crystalline alpha cellulose fraction of the cellulose the cellulose is impregnated with acid at a concentration of 0.05% to 1.0% of the weight of the cellulose, and the pressure vessel is rapidly filled with steam to a pressure between 2.41 to 3.79 MPa (350 and 550 psi) to bring the acid-impregnated cellulose to a temperature in the range of 200—225°C in less than 60 seconds and then explosively expelled to the atmosphere.

If the desired product is a mixture of glucose and level off DP microcrystalline alpha cellulose, it is preferred to use hydrochloric acid to impregnate the cellulose and to treat the cellulose with steam at pressures in the range of 2.07 to 3.10 MPa (300 to 450 psi). If on the other hand, the desired product is substantially pure glucose, it is preferred to use sulfuric acid to impregnate the cellulose and to treat the cellulose with steam at pressures in the range of 2.76 to 4.82 MPa (400 to 700 psi).

In some embodiments of the present invention wherein the neutralised end product is a mixture of glucose from the amorphous component of the cellulose and level off DP microcrystalline alpha cellulose from the crystalline alpha cellulose fraction of the cellulose, the cellulose is impregnated with hydrochloric acid at a concentration of about 0.2% of the weight of the cellulose, and the pressure vessel is rapidly filled with steam to a pressure of about 3.10 MPa (450 psi) to bring the acid impregnated cellulose to a temperature of about 215°C in less than 45 seconds and then explosively expelled to the atmosphere. In yet other embodiments of the present invention wherein the end product is substantially all glucose, the pressure vessel is rapidly filled with steam to a pressure in the range 2.76 to 4.82 MPa (400 to 700 psi), to bring the cellulose, which has been impregnated with sulfuric acid to a level in the range 0.5% to 1.5% of the weight of the cellulose, to a temperature in the range 215 to 240°C in less than 60 seconds and then explosively expelled to the atmosphere.

In other embodiments of the present invention wherein the end product is substantially all glucose, the pressure vessel is rapidly filled with steam to a pressure of about 4.48 MPa (650 psi) to bring the cellulose, which has been impregnated with sulphuric acid to a level of about 1.0% of the weight of the cellulose, to a temperature of about 234° in less than 45 seconds. The contents of the pressure vessel are then explosively expelled to the atmosphere.

Preferably, condensate, which is produced when the high pressure steam contacts the relatively cool cellulosic material, is removed from the bottom of the pressure vessel as it is formed.

Any residual cellulose may be treated with enzymes for the conversion thereof to glucose or retreated in the pressure vessel. In the accompanying drawings which illustrates an embodiment of the invention,

Figure 1 is a sectional side view of a pressure vessel having a valved outlet,

In Figure 1, there is shown a pressure vessel 2, having a valved outlet which in the embodiment illustrated is an extrusion die outlet 6, an extrusion die closure plug 30, a loading end closure flaps 8, and steam inlet orifices 10 to 12. The pressure vessel 2 has a bottleneck portion 14 leading to the die 4 and entry ports 16 and 18 for temperature probes (not shown).

The front end of the pressure vessel 2, containing the die outlet 14, has a flange 20 to which is sealed a curved impinging tube 22 which gradually reduces in cross-section in a downstream direction. The curved impinging tube 22 has a spindle inlet sleeve 24 provided with a flange 26. A pneumatic ram 28 is attached to the flange 26 and has a die closure plug 30 mounted on the spindle 32 of the ram 28. A condensate drainage tank 31 is provided having an outlet valve 33, thus permitting the removal of condensate as it is formed.

The rear end 34 of the pressure vessel 2 is sealed to the remainder by flanges 36 and 38 and has the loading end closure flap 8 hinged thereto by a hinge 40 and is sealable therewith by a clamp 42. The rear end 34 has a venting safety valve 44.

The pressure vessel shown in Figure 1 may be used in the process described herein both in the initial step of pressurizing and explosively expelling the divided lignocellulosic material, and in the subsequent step of pressurizing and explosively expelling the acid-impregnated cellulose fibrils.

In operation the loading end closure flap 8 is opened and the pressure vessel 2 is loaded with lignocellulosic material in a divided form with the die closure plug 30 closing the die outlet 4. A rod (not shown) is used to pack the lignocellulosic material in the pressure vessel 2.

With the pressure vessel 2 completely filled with lignocellulosic material the die closure plug 30 is sealed by the pneumatic ram 28 and the closure plug 8 is sealed to the rear end 34 by the clamp 42 and then the pressure vessel is filled with steam at a pressure in the range 2.76 to 4.82 MPa (400 to 700 psi), and at a sufficient temperature to raise the temperature of the lignocellulosic material to a temperature in the range 185 to 240°C, in less than 60 seconds to thermally soften the lignocellulosic material into a plastic by injecting steam into the steam inlet orifices 10 to 12 from a source (not shown). The temperature probes (not shown) in the ports 16 and 18 are used to monitor the temperature of the lignocellulosic material in the pressure vessel 2 to determine when the lignocellulosic material has reached the chosen temperature.

As soon as the lignocellulosic material in the pressure vessel 2 reaches the desired temperature the pneumatic ram 28 is actuated to withdraw the closure plug 30 and more or less instantaneously open the die outlet 4 to atmosphere so that the lignocellulosic material is extruded through the die outlet 4 in the plasticized

condition and at the extrusion pressure and is flashed to atmosphere preferably in milli-seconds along the curved impinging tube 22. Thus sudden release to atmosphere explosively expels the lignocellulosic material in the plasticized condition and produces a particulate material having the appearance of potting soil which stains the fingers brown and has a high enough specific gravity to sink like a stone in water.

While the curved impinging tube is not essential it has the advantage of utilizing some of the extrusion force to further comminute the lignocellulosic material in addition to the comminution obtained by extrusion.

The cellulose is then separated from the particulate product, by the methods described above. It is then soaked in a solution of sulfuric, hydrochloric or sulfurous acid and the moisture content reduced.

The loading end closure flap 8 of the pressure vessel 2 is then opened and the pressure vessel is loaded with the acid-impregnated cellulose with the die closure plug 30, and the valve 33 closed.

With the pressure vessel 2 completely filled with acid-impregnated cellulose, the die closure plug 30 is sealed by the pneumatic ram 28 and the closure plug 8 is sealed to the rear end 34, by the clamp 42, and then the pressure vessel is filled with steam at a pressure of 2.07 MPa (300 psi) to 4.82 MPa (700 psi), to bring the cellulose to a temperature between 185 and 240°C in less than 60 seconds, preferably less than 45 seconds, depending on the moisture content of the material and the pH of the acid solution impregnated into the cellulose. The temperature probes (not shown) in the ports 16 and 18 are used to monitor the temperature of the acid-impregnated cellulose to determine when the cellulose has reached the chosen temperature.

As soon as the cellulose in the pressure vessel has reached the desired temperature, the valved outlet is opened and the material is explosively expelled from the pressure vessel through the outlet to atmosphere.

The percentage of acid used in the impregnation step will depend on the temperature to be used, the time to achieve that temperature and the moisture content of the impregnated cellulose. In any case, acid concentration will be no more than two percent of the dry weight of the material, and usually much less. The object of the process is to weaken, by acid hydrolysis and heat, the intramolecular bonds joining the glucose units to form the cellulose. The combination of the acid hydrolysis and the mechanical shock caused by the instant decompression and the expulsion through the outlet will fracture those bonds, producing a high concentration of glucose and at the same time reducing the pressure to atmosphere, thus reducing the temperature to below 100°C which will quench further chemical hydrolysis.

The resulting product can then be neutralized by a suitable base and filtered to remove the residual cellulose which can be used as described

above or returned to the pressure vessel for a second treatment, or passed to an enzymatic hydrolysis process to complete the conversion to glucose.

During the first few seconds after the steam is introduced into the reactor, contact with the relatively cool cellulosic material produces a liquid condensate. This condensate covers between 10 and 30 percent of the cellulosic material depending on the moisture content and the starting temperature of the cellulose feedstock, thus preventing proper processing of the submerged cellulosic material. The tank 31 for removing that condensate, as it is formed, substantially improves the performance of the process.

Claims

- 20 1. A method of dissociating lignocellulosic material, comprising:
 - a) packing the lignocellulosic material in a divided, exposed, moist form in a pressure vessel having a valved outlet;
 - b) with the valve closed, rapidly filling the pressure vessel with steam at high pressure to bring, by means of the pressurised steam, substantially all of the lignocellulosic material to a predetermined temperature in the range 185 to 240°C in less than 60 seconds;
 - c) as soon as the said predetermined temperature has been attained, opening the valved outlet and explosively expelling the lignocellulosic material in a plastic condition from the pressure vessel through the outlet to atmosphere so that the said material issues from the outlet in particulate form and;
 - d) separating the cellulose in the particulate material from the lignin and hemicellulose, characterised by said high pressure steam being at a pressure of 2.76 to 4.82 MPa (400 to 700 psi) and by the further steps of:
 - e) soaking the separated cellulose in an acid solution of at least one acid selected from the group consisting of sulfuric acid, hydrochloric acid and sulfurous acid until the acid is distributed evenly throughout the cellulose, and then reducing the moisture content of the cellulose, leaving the cellulose impregnated with acid at a concentration in the range 0.05% to 2.0% of the weight of the cellulose;
 - f) packing the acid-impregnated cellulose into a pressure vessel having a valved outlet;
 - g) rapidly filling the pressure vessel with steam at a pressure in the range 2.07 to 4.82 MPa (300 psi to 700 psi) to bring, by means of the pressurised steam, substantially all of the acid-impregnated cellulose to a predetermined temperature in the range of 185—240°C in less than 60 seconds;
 - h) as soon as the said predetermined temperature has been attained, opening the valved outlet, and explosively expelling the acid-impregnated cellulose from the pressure vessel through the outlet to atmospheric pressure;

- i) neutralising the resulting solution.
2. A method according to claim 1, wherein any residual cellulose is filtered from said solution.
3. A method according to claim 1 or claim 2, wherein the acid-impregnated cellulose is reduced to a moisture content of the level in the range of 20% to 80% by weight of the cellulose before being packed into the pressure vessel.
4. A method according to any preceding claim, wherein the end product is a mixture of glucose from the amorphous component of the cellulose and level off DP microcrystalline cellulose from the crystalline alpha cellulose fraction of the cellulose, and in step (e) the cellulose is impregnated with acid at a concentration of 0.05% to 1.0% of the weight of the cellulose, and in step (g) the pressure vessel is rapidly filled with steam to a pressure between 2.41 to 3.79 MPa (350 psi and 550 psi) to bring the acid-impregnated cellulose to a temperature in the range of 200°C to 225°C in less than 60 seconds.
5. A method according to any of claims 1 to 3, wherein the end product is a mixture of glucose from the amorphous component of the cellulose and level off DP microcrystalline alpha cellulose from the crystalline alpha cellulose fraction of the cellulose, and in step (e) the cellulose is impregnated with hydrochloric acid at a concentration of about 0.2% of the weight of the cellulose, and in step (g) the pressure vessel is rapidly filled with steam to a pressure of about 3.10 MPa (450 psi) to bring the acid-impregnated cellulose to a temperature of about 215°C in less than 45 seconds.
6. A method according to any of claims 1 to 3, wherein the end product is substantially all glucose, in step (g) the pressure vessel is rapidly filled with steam to a pressure in the range 2.76 to 4.82 MPa (400 psi to 700 psi), to bring the cellulose, which has been impregnated with sulphuric acid to a level in the range 0.5% to 1.5% of the weight of the cellulose, to a temperature in the range of 215°C to 240°C, in less than 60 seconds.
7. A method according to any of claims 1 to 3, wherein the end product is substantially all glucose, in step (g) the pressure vessel is rapidly filled with steam to a pressure of about 4.48 MPa (650 psi) to bring the cellulose, which has been impregnated with sulphuric acid to a level of about 1% of the weight of the cellulose, to a temperature of about 234°C, in less than 45 seconds.
8. A method according to any preceding claim, wherein liquid condensate is removed from the bottom of the pressure vessel as it is formed.
9. A method according to any preceding claim, wherein steps (e) to (i) are repeated on any residual cellulose.
10. A method according to any of claims 1 to 8 wherein any residual cellulose is treated with enzymes for the conversion thereof to glucose.
- Patentansprüche**
1. Verfahren zum Spalten von Holzzellstoffmaterial, bei welchem:
- a) das Holzzellstoffmaterial in zerteilter, expo-
- nierter, feuchter Form in einen Druckkessel mit einem ventilbeherrschten Auslaß gepackt wird;
- b) der Druckkessel bei geschlossenem Ventil schnell mit Dampf hohen Druckes gefüllt wird, um mittels des Druckdampfes im wesentlichen das gesamte Holzzellstoffmaterial auf eine vorbestimmte Temperatur im Bereich von 185 bis 240°C in weniger als 60 s zu bringen;
- c) der ventilbeherrschte Auslaß geöffnet wird, sobald die vorbestimmte Temperatur erreicht worden ist, und das Holzzellstoffmaterial, explosionsartig in plastischem Zustand durch den Auslaß aus dem Druckkessel in die Atmosphäre ausgebracht wird, so daß das Material aus dem Auslaß in Teilchenform austritt, und
- d) die Zellulose in dem Teilchenmaterial von dem Lignin und der Hemizellulose getrennt wird, dadurch gekennzeichnet, daß der Hochdruckdampf sich auf einem Druck von 2,76 bis 4,82 MPa (400 bis 700 psi) befindet und daß:
- e) die abgetrennte Zellulose mit einer Säurelösung aus wenigstens einer Säure, die aus der Gruppe bestehend aus Schwefelsäure, Chlorwasserstoffsäure und schwefeliger Säure ausgewählt ist, durchtränkt wird, bis die Säure in der Zellulose gleichmäßig verteilt ist, und dann der Feuchtigkeitsanteil der Zellulose verringert wird, bis die Zellulose mit Säure in einer Konzentration im Bereich von 0,05% bis 2,0% des Gewichtes der Zellulose imprägniert ist;
- f) die mit Säure imprägnierte Zellulose in einen Druckkessel mit einem ventilbeherrschten Auslaß gepackt wird;
- g) der Druckkessel schnell mit Dampf mit einem Druck in dem Bereich von 2,07 bis 4,82 MPa (300 psi bis 700 psi) gefüllt wird, um im wesentlichen die gesamte mit Säure imprägnierte Zellulose mittels des Druckdampfes auf eine vorbestimmte Temperatur im Bereich von 185 bis 240°C in weniger als 60 s zu bringen;
- h) der ventilbeherrschte Auslaß geöffnet wird, sobald die vorbestimmte Temperatur erreicht worden ist, und die mit Säure imprägnierte Zellulose aus dem Druckkessel durch den Auslaß explosionsartig auf atmosphärischen Druck gebracht wird;
- i) die sich ergebende Lösung neutralisiert wird.
2. Verfahren nach Anspruch 1, bei welchem jede Restzellulose aus der Lösung ausgefiltert wird.
3. Verfahren nach Anspruch 1 oder 2, bei welchem der Feuchtigkeitsanteil der mit Säure imprägnierten Zellulose auf einen Wert im Bereich von 20% bis 80% des Gewichtes der Zellulose verringert wird, bevor die imprägnierte Zellulose in den Druckkessel gepackt wird.
4. Verfahren nach einem der vorhergehenden Ansprüche, bei welchem das Endprodukt ein Gemisch aus Glukose der amorphen Komponente der Zellulose und DP-Mikrokristallinzellulose aus der kristallinen Alphazellulosefraktion der Zellulose ist und bei welchem im Schritt (e) die Zellulose mit Säure einer Konzentration von 0,05% bis 1,0% des Gewichtes der Zellulose imprägniert wird und im Schritt (g) der Druckkessel schnell mit Dampf bis zu einem Druck zwischen 2,41 bis 3,79

MPa (350 psi bis 550 psi) gefüllt wird, um die mit Säure imprägnierte Zellulose auf eine Temperatur im Bereich von 200°C bis 225°C in weniger als 60 s zu bringen.

5. Verfahren nach einem der Ansprüche 1 bis 3, bei welchem das Endprodukt ein Gemisch aus Glukose aus der amorphen Komponente der Zellulose und DP-mikrokristallin-Alphazellulose aus der kristallinen Alpha-zellulosefraktion der Zellulose ist und bei welchem die Zellulose im Schritt (e) mit Chlorwasserstoffsäure in einer Konzentration von etwa 0,2% des Gewichtes der Zellulose imprägniert wird und der Druckkessel im Schritt (g) mit Dampf bis zu einem Druck von etwa 3,10 MPa (450 psi) schnell gefüllt wird, um die mit Säure imprägnierte Zellulose auf eine Temperatur von etwa 215°C in weniger als 45 s zu bringen.

6. Verfahren nach einem der Ansprüche 1 bis 3, bei welchem das Endprodukt im wesentlichen vollständig Glukose ist und bei welchem im Schritt (g) der Druckkessel rasch mit Dampf bis zu einem Druck im Bereich von 2,76 bis 4,82 MPa (400 psi bis 700 psi) gefüllt wird, um die Zellulose, die mit Schwefelsäure auf einen Grad im Bereich von 0,5% bis 1,5% des Gewichtes der Zellulose imprägniert wurde, auf eine Temperatur im Bereich von 215 bis 240°C in weniger als 60 s zu bringen.

7. Verfahren nach einem der Ansprüche 1 bis 3, bei welchem das Endprodukt im wesentlichen vollständig Glukose ist und bei welchem im Schritt (g) der Druckkessel schnell mit Dampf bis zu einem Druck von etwa 4,48 MPa (650 psi) gefüllt wird, um die Zellulose, welche mit Schwefelsäure bis zu einem Grad von 1% des Gewichtes der Zellulose imprägniert wurde, auf eine Temperatur von etwa 234°C in weniger als 45 s zu bringen.

8. Verfahren nach einem der vorangehenden Ansprüche, bei welchem Flüssigkondensat bei dessen Bildung vom Boden des Druckkessels entfernt wird.

9. Verfahren nach einem der vorangehenden Ansprüche, bei welchem die Schritte (e) bis (i) für jede Restzellulose wiederholt werden.

10. Verfahren nach einem der Ansprüche 1 bis 8, bei welchem jede Restzellulose für deren Umwandlung in Glukose mit Enzymen behandelt wird.

Revendications

1. Un procédé de dissociation de matériau lignocellulosique comprenant:

a) la compression du matériau lignocellulosique à l'état divisé brut et humide dans un récipient sous pression ayant une sortie commandée par une vanne;

b) la vanne étant fermée, le remplissage rapide du récipient sous pression avec de la vapeur à haute pression pour amener, au moyen de la vapeur sous pression, pratiquement tout le matériau lignocellulosique à une température préterminée de l'ordre de 185 à 240°C en moins de 60 secondes;

5 c) dès que ladite température préterminée a été atteinte, l'ouverture de la sortie commandée par vanne et l'expulsion explosive du matériau lignocellulosique à l'état plastique par la sortie du récipient sous pression vers l'atmosphère de sorte que ledit matériau sort sous forme particulaire; et

10 d) la séparation de la cellulose dans le matériau particulaire, de la lignine et l'hémicellulose, caractérisé par le fait que la haute pression de vapeur est à une pression de 2,76 à 4,82 MPa (400 à 700 psi) et par les étapes suivantes consistant en:

15 e) le trempage de la cellulose séparée dans une solution acide d'au moins un acide choisi dans le groupe consistant en l'acide sulfurique, l'acide chlorhydrique et l'acide sulfureux jusqu'à ce que l'acide soit réparti régulièrement dans toute la cellulose, la teneur en humidité de la cellulose étant alors réduite, laissant la cellulose imprégnée d'acide à une concentration de l'ordre de 0,05% à 2,0% en poids de la cellulose;

20 f) la compression de la cellulose imprégnée d'acide dans un récipient sous pression ayant une sortie commandée par vanne;

25 g) le remplissage rapide du récipient sous pression avec de la vapeur à une pression de l'ordre de 2,07 à 4,82 MPa (300 à 700 psi) pour amener, au moyen de la vapeur sous pression, pratiquement toute la cellulose imprégnée d'acide à une température préterminée de l'ordre de 185 à 240°C en moins de 60 secondes;

30 h) dès que ladite température préterminée est atteinte, l'ouverture de la vanne et l'expulsion explosive par la sortie de la cellulose imprégnée d'acide du récipient sous pression vers la pression atmosphérique;

i) la neutralisation de la solution résultante;

35 2. Un procédé selon la revendication 1, dans lequel toute la cellulose résiduelle est filtrée de ladite solution.

3. Un procédé selon la revendication 1 ou la revendication 2 dans lequel la cellulose imprégnée d'acide est réduite à une teneur en humidité d'un niveau de l'ordre de 20 à 80% en poids de cellulose avant d'être comprimé dans le récipient sous pression.

40 4. Un procédé selon l'une quelconque des revendications précédentes, dans lequel le produit final est un mélange de glucose du composant amorphe de la cellulose, et de la cellulose microcristalline à degré de polymérisation hors norme de la fraction d'alpha cellulose cristalline de la cellulose et dans lequel dans l'étape e) la cellulose est imprégnée avec un acide à une concentration de 0,05% à 1,0% en poids de la cellulose et dans l'étape g) le remplissage rapide du récipient sous pression est effectué avec de la vapeur à une pression comprise entre 2,41 et 3,79 MPa (350 à 550 psi) pour amener la cellulose imprégnée d'acide à une température de l'ordre de 200 à 225°C en moins de 60 secondes.

45 5. Un procédé selon l'une quelconque des revendications 1 à 3, dans lequel le produit final est un mélange de glucose du composant

amorphe de la cellulose et d'alphacellulose monocristalline à degré de polymérisation hors norme de la fraction d'alphacellulose cristalline de la cellulose, et dans lequel dans l'étape e) la cellulose est imprégnée avec de l'acide chlorhydrique à une concentration d'environ 0,2% en poids de la cellulose, et dans l'étape g) le remplissage rapide du récipient sous pression s'effectue avec de la vapeur à une pression d'environ 3,10 MPa (450 psi) pour amener la cellulose imprégnée d'acide à une température d'environ 215°C en moins de 45 secondes.

6. Un procédé selon l'une quelconque des revendications 1 à 3, dans lequel le produit final est pratiquement tout du glucose, dans lequel dans l'étape g) le remplissage rapide du récipient sous pression est effectué avec de la vapeur à une pression de l'ordre de 2,76 à 4,82 MPa (400 à 700 psi) pour amener la cellulose qui a été imprégnée avec de l'acide sulfurique à un niveau de l'ordre de 2,05% à 1,5% en poids de la cellulose, à une température de l'ordre de 215 à 240°C en moins de 60 secondes.

7. Un procédé selon l'une quelconque des revendications 1 à 3, dans lequel le produit final est pratiquement tout du glucose et dans lequel dans l'étape g) le remplissage rapide du récipient sous pression est effectué avec de la vapeur à une pression d'environ 4,48 MPa (650 psi) pour amener la cellulose, qui a été imprégnée avec de l'acide sulfurique à un niveau d'environ 1% en poids de la cellulose, à une température de 234°C en moins de 45 secondes.

8. Un procédé selon l'une quelconque des revendications précédentes, dans lequel le condensat est extrait du fond du récipient sous pression quand il est formé.

9. Procédé selon l'une quelconque des revendications précédentes, dans lequel les étapes e), a), i) sont répétées sur toute cellulose résiduelle.

10. Procédé selon l'une quelconque des revendications 1 à 8, dans lequel toute cellulose résiduelle est traitée avec des enzymes pour sa conversion en glucose.

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